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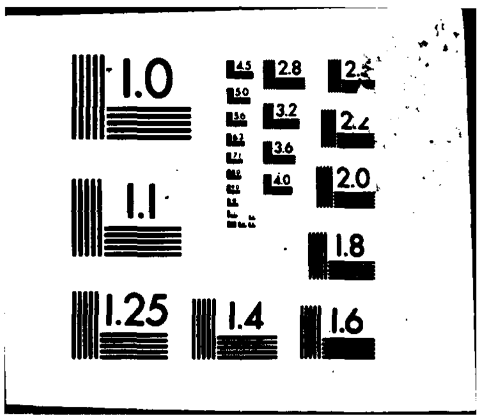
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EPITAXIAL GARNETS AND HEXAGONAL FERRITES

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The objective of this research is to develop new and improved epitaxial ferrite materials for use in microwave and millimeter-wave signal processing devices. The major emphasis has been on multiple layer magnetic garnet structures for magnetostatic wave (MSW) delay lines. Theoretical analysis and experiments were performed on propagation of magnetostatic surface waves (MSSW), magnetostatic forward volume waves (MSFVW) and magnetostatic backward volume waves (MSBVW) in structures containing up to four layers. In the experiments, one layer was a standard stripline substrate, 10 mil thick alumina. The other

Abstract: (Cont.)

layers were YIG (yttrium iron garnet) films grown by liquid phase epitaxy (LPE) on gadolinium gallium garnet (GGG) substrates. Magnetic properties of the YIG layers were adjusted by modifying their chemical composition. Delay characteristics were measured in detail at frequencies between 2 and 4 GHz. Using MSSW, a multilayer structure having a flatter nondispersive delay than conventional single layer devices was demonstrated. Linear dispersion similar to that of conventional single layer devices but easier to implement was demonstrated for MSFVW propagation in a structure containing three epitaxial layers. In addition to the MSW work on garnets, LPE of lithium ferrite and hexagonal ferrites was studied. A substituted lead hexaferrite which exhibited 50 GHz resonance in a field of only 3 KOe was demonstrated.

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EPITAXIAL GARNETS AND HEXAGONAL FERRITES

AFOSR Contract F49620-80-C-0045

Final Technical Report

H. L. Glass and L. R. Adkins, Principal Investigators

April 20, 1982

I. OBJECTIVES

The overall objective of this research is to develop epitaxial single crystal ferrite films suitable for microwave and millimeter-wave signal processing at frequencies above 1 GHz. The specific tasks are:

a. Analyze and develop special layered garnet structures for propagating-magnetic-wave devices operating at frequencies between 1 GHz and 25 GHz.

b. Investigate the use of multichannel stacks to produce wide bandwidth nondispersive MSW behavior.

c. Investigate LPE growth of lithium ferrite with the objective of preparing low-loss, large area films suitable for delay lines.

d. Investigate epitaxial hexagonal ferrites for use in devices operating at frequencies above 70 GHz, including investigation of substitutions such as Ga or Al as a method for raising anisotropy fields to produce materials useful for devices operating at frequencies above 100 GHz.

These tasks represent a continuation of the work done under previous contracts, F49620-79-C-0048 and F44620-75-C-0045. Tasks a. and b. relate to magnetostatic wave (MSW) devices similar to surface acoustic wave (SAW) devices, but operating at higher frequencies. These tasks make use of the fairly well established epitaxial garnet technology. Tasks c. and d. represent an exploration of the epitaxial growth of other classes of ferrites which have capabilities beyond those of the garnet-structure ferrites.

II. ACCOMPLISHMENTS

Of the four tasks, Task a. has been given the highest priority and considerable progress has been made. Task b. can be considered as a specific application of Task a. An important factor in our planning of work on these two tasks has been the need to determine whether delay line type devices based on magnetostatic wave propagation can exhibit the dispersion control

and bandwidth needed for practical applications. Tasks c. and d. are higher risk activities aimed at producing epitaxial films of lithium ferrite and hexagonal ferrite of a quality suitable for both device development and research on fundamental properties of these materials.

a. A theoretical model was developed to calculate the propagation characteristics of magnetostatic waves in multilayer media consisting of up to four layers. In practice, one layer is the standard 10 mil thick alumina substrate of a stripline. The other layers are epitaxial films, generally YIG (yttrium iron garnet) with magnetic properties adjusted by suitable modifications of composition. The epitaxial layers are grown on a GGG (gadolinium gallium garnet) substrate crystal. The model was applied to calculations of all three principal modes of propagation: MSSW (magnetostatic surface waves), MSFVW (magnetostatic forward volume waves) and MSBVW (magnetostatic backward volume waves) for realistic ranges of layer magnetization and thickness.

The results of the theoretical analysis were used to set target values to guide growth of the epitaxial YIG films. Aluminum or gallium substitutions for iron were used in combination with lanthanum substitutions for yttrium in order to achieve desired magnetization values while maintaining lattice matching to the GGG substrates. These substitutions have the effect of reducing the magnetization of the YIG even to the point at which the layer can serve as a nonmagnetic dielectric. A large number of single, double and triple epitaxial layers were prepared to test the theoretical predictions. The layers were characterized by FMR (ferromagnetic resonance) and by magnetostatic wave delay line measurements for the three principal modes. There was excellent agreement between measured dispersive characteristics and the predictions of the theoretical model.

After experimental verification, the model was used to devise multilayer structures which would exhibit greater linearity in linearly dispersive delay or greater flatness in nondispersive delay while maximizing bandwidth. These structures were grown and tested. Results showed that the nondispersive behavior for MSSW could be made much flatter than for conventional single layer delay lines. This result was demonstrated in structures which contained two magnetic layers. Details are given in items 1, 2 and 4 in the list of publications. For linearly dispersive delay, best results were obtained when the structure contained two magnetic layers having nearly the same magnetization but separated by a thin (~25μm) nonmagnetic layer. Details are given in items 4 and 6 in the list of publications. The latter reference shows that the triple layer delay line offers several advantages over single layer delay lines. In particular, the triple layer line is actually easier to fabricate and is compatible with conventional transducer and planar waveguide structures.

b. The multichannel stack is a concept for increasing the bandwidth of a nondispersive delay line. In its simplest form, the stack would contain two delay lines. The two lines would be fabricated using slightly different YIG compositions so that they would differ in magnetization. Then, when placed in a common applied magnetic field their passbands would be displaced to provide a total passband nearly double that of a single delay line. In principle, several such delay lines could be combined in a single package to obtain bandwidths several times that of a single line.

Our initial efforts included the growth of films of different YIG composition on opposite faces of a substrate wafer to provide a monolithic pair of delay lines. Results obtained by feeding each delay line separately were reported in item 2 in the list of publications. Simultaneous feed was accompanied by high feedthrough which prevented a reliable quantitative comparison with the theoretical calculations. Subsequently, additional samples were prepared and a new package was designed. Feedthrough was reduced substantially. However, insertion loss was high. Definitive testing of the stack will require design and fabrication of external circuits to provide better matching between channels. This is beyond the scope of this contract, but would be appropriate for a device-oriented program. We have, in fact, incorporated some aspects of the multichannel stack into a proposal to RADC for development of electronically variable time delay elements for a phased array radar system. This proposal also drew upon the extensive theoretical analyses developed on this contract.

c. Our lithium ferrite work was a direct continuation of the work done on the previous contracts. This included characterization of materials prepared under those contracts. These results were reported; item 3 in the list of publications and items 2 and 4 in the list of oral presentations. In that work, the best films (best in terms of smoothness, uniformity and absence of extra modes in the FMR spectra) were prepared at a temperature of 875°C from a melt which contained Fe_2O_3 and Li_2CO_3 in mole ratio 3.9:1 and having a solute concentration of 18.3 mole % in a $PbO:B_2O_3$ flux having mole ratio 28.6:1.

Subsequently, we tried to find a modified melt composition which would permit growth at lower temperatures. We anticipated several advantages: lower thermal stress, reduced interdiffusion between film and substrate, and the possibility of growth at a temperature below that of the order-disorder transition. To accomplish this we kept the $PbO:B_2O_3$ ratio at about 28.6:1 but reduced the solute concentration to 9.4 mole % while increasing the iron excess, Fe_2O_3: Li_2CO_3, to 5.4:1. This melt froze without formation of ferrite, so the Fe_2O_3 content was gradually increased. As this was done, we found that $LiFeO_2$ was the primary crystallizing species until the Fe_2O_3 content was increased to provide a solute concentration of 15.2 mole % and an Fe_2O_3:Li_2CO_3 ratio of 10.0:1 at which point the desired $LiFe_5O_8$ phase

became the primary crystallizing species. Films were then grown at temperatures in the range from 871 to 884°C on magnesium indium-gallate spinel substrates. There was no difficulty with nucleation in the melt and film quality appeared to be similar to that observed previously; smooth films on (111) substrates but some growth terraces, rough films on (100) substrates. Microwave characterization of these films is planned for the near future. Obviously, however, this melt composition did not enable us to reduce the growth temperature by any significant amount. It may be worthwhile to try further modifications to the melt composition to reduce the growth temperature.

d. During the first year of this contract, the hexagonal ferrite task included frequencies as low as 25 GHz. This was modified to frequencies above 70 GHz because of the increased interest in higher frequencies and to avoid duplicating the effort planned for a new program which we began in November 1980 under contract to the Army Research Office. The Army program, "Epitaxial Hexagonal Ferrites for Millimeter-wave Tunable Filters," is aimed at applications in the 50 GHz to 70 GHz frequency range. A major part of the Army program is to improve the crystal quality of the epitaxial hexagonal ferrites since this is believed to be necessary for attainment of lower losses. With the expectation that this contract effort would benefit from improvements in quality developed on the Army program, we deferred our planned activities at the higher frequencies.

The hexagonal ferrite work which was performed during this contract was focussed on PbO-based hexagonal ferrites. We succeeded in growing the M-type lead hexaferrite (magnetoplumbite) on gallate spinel substrates. We found that the PbO-based hexaferrite was easier to grow than the BaO-based hexaferrites which we grew under the previous contracts. However, FMR linewidths were a little higher. Also, the PbO-based hexaferrite has a somewhat lower anisotropy field so that slightly higher external fields are required for resonance. We were able to demonstrate that the anisotropy field could be increased by substitution of Ga for some of the Fe atoms and we observed 50 GHz resonance at an applied field of 3 KOe. For unsubstituted BaO-based hexaferrite, a field of 5 KOe would be necessary.

The results we have obtained on the PbO-based hexagonal ferrites have been reported, see item 3 in the list of publications and items 2 - 6 in the list of oral presentations. This work also served as the basis for two patents, see items 7 and 8 in the list of patents issued.

In our Army program, we are growing SrO-based hexagonal ferrites from a PbO-based flux. Thus, the ferrites are actually mixed Pb, Sr hexaferrites. The substrates are gallate spinel crystals. The substrate growth techniques and the hexagonal ferrite liquid phase epitaxy techniques are direct derivatives from this contract and the previous contracts.

III. PERSONNEL

The Principal Investigators were H. L. Glass, who carried out most of the crystal growth and crystallographic characterization, and L. R. Adkins, who performed the FMR and microwave analyses. F. S. Stearns contributed to the crystal growth and crystal processing activities.

IV. PUBLICATIONS

1. L. R. Adkins and H. L. Glass, "Propagation of Magnetostatic Surface Waves in Multiple Magnetic Layer Structures," Electron. Lett. 16 (1980) 590-592.

2. L. R. Adkins and H. L. Glass, "Dispersion Control in Magnetostatic Delay Lines by Means of Multiple Magnetic Layer Structures," Proc. IEEE 1980 Ultrasonics Symposium, 80CH1602, 526-531.

3. H. L. Glass, F. S. Stearns and L. R. Adkins, "LPE Growth of Magnetoplumbite and Lithium Ferrite from $PbO-B_2O_3$ Flux," in FERRITES: Proc. 3rd Int. Conf. on Ferrites, Japan (1980) 39-42.

4. L. R. Adkins and H. L. Glass, "Nondispersive and Linearly Dispersive MSW Propagation on Multilayer Films," Proceedings RADC Microwave Magnetics Technology Workshop, Hanscom AFB (1981) in press.

5. H. L. Glass, "Epitaxial Ferrites for Magnetostatic Wave Devices," Proceedings RADC Microwave Magnetics Technology Workshop, Hanscom AFB (1981) in press.

6. L. R. Adkins and H. L. Glass, "Magnetostatic Volume Wave Propagation in Multiple Ferrite Layers," J. Appl. Phys. submitted.

V. SCIENTIFIC INTERACTIONS

Work performed under this contract has been reported in several oral presentations. Four of these presentations have been included in published conference proceedings and were listed in the preceding section. The following is a list of oral presentations for which no proceedings will be published. Also included in this list are two planned presentations.

1. H. L. Glass and V. S. Speriosu, "Tailoring Magnetic Properties in Epitaxial Ferrites," U.S. - France Seminar on Applications of X-ray Topography to Materials Science, University of Paris, March 1980.

2. H. L. Glass and F. S. Stearns, "LPE Growth of Ferrites from $PbO-B_2O_3$ Flux," 5th Conf. on Crystal Growth, Amer. Assoc. for Crystal Growth Western Section, Fallen Leaf Lake, May 1980.

3. H. L. Glass and L. R. Adkins, "Epitaxial Single Crystal Thin Film Ferrites," Workshop on Millimeter Wave Devices Using Gyrotropic Media, IEEE/MTT-S International Microwave Symposium, Washington, D.C. May 1980.

4. H. L. Glass, "Liquid Phase Epitaxy of Hexagonal and Spinel Ferrites," INTERMAG Conference, Grenoble, May 1981.

5. H. L. Glass, ARO Workshop on Short Millimeter Wave Nonreciprocal Materials and Devices, Army Research Office, Research Triangle Park, Nov. 1981.

6. F. S. Stearns and H. L. Glass, "Growth of Large Gallate Spinel Crystals from Lead Oxide - Boron Oxide Flux," 6th Conf. on Crystal Growth Amer. Assoc. for Crystal Growth Western Section, Fallen Leaf Lake, June 1982, submitted.

7. L. R. Adkins and H. L. Glass, "Forward and Backward Magnetostatic Volume Modes in Multiple Layer Ferrite Films," 3rd Joint INTERMAG - Magnetism and Magnetic Materials Conf., Montreal, Jul. 1982, submitted.

8. H. L. Glass, Magnetostatic Wave Device Technology Workshop, 3rd Joint INTERMAG - Magnetism and Magnetic Materials Conf., Montreal, Jul. 1982.

Presentations number 1, 3, 4, 5 and 8 on this list and the presentation of number 5 in the list of publications were invited talks.

Our interactions during the contract period included meetings and discussions with researchers from a number of universities, companies and government laboratories. Some of these interactions took place at conferences and workshops. In addition, there were several visits to these researchers' laboratories or visits by them to our laboratory. The government laboratories with which we interacted included RADC, AFWAL, NRL and ERADCOM. These interactions mainly involved informal briefings to keep government technical personnel informed of our research progress and to discuss potential applications of our work to military electronic systems. Samples of our epitaxial ferrite materials were provided to NRL and ERADCOM to help support some of their in-house research activities.

University and company interactions included researchers working on microwave ferrites and magnetostatic wave devices at MIT, Georgia Tech, University of Texas, University of California at Irvine, Colorado State University, State University of New York at Stony Brook, Carnegie-Mellon University, Osaka University, Osaka Electro-Communication University, Westinghouse, Litton and Philips Research Laboratories. We supplied samples of our ferrite materials to Georgia Tech, for their studies on ion-beam modification of

magnetic properties, to UC Irvine, for their studies on optical interactions with magnetostatic waves, to Colorado State University, for their fundamental studies on microwave interactions with ferrites, and to SUNY Stony Brook, for their studies on magnetostatic wave devices.

VI. INVENTIONS

During the period of this contract, five invention disclosures have been filed with our company patent department. Some of these involved work performed under the previous contracts. The inventions have been reported separately in Abstracts of New Technology submitted to AFOSR.

The following United States Patents were issued during the contract period:

1. 4,189,521 "Epitaxial Growth of M-Type Hexagonal Ferrite Films on Spinel Substrates and Composite."

2. 4,200,484 "Method of Fabricating Multiple Layer Composite."

3. 4,243,697 "Self Biased Ferrite Resonators."

4. 4,263,374 "Temperature-Stabilized Low-Loss Ferrite Films."

5. 4,269,651 "Process for Preparing Temperature-Stabilized Low-Loss Ferrite Films."

6. 4,273,610 "Method for Controlling the Resonance Frequency of Yttrium Iron Garnet Films."

7. 4,292,119 "Growth of Single-Crystal $2PbO \cdot Fe_2O_3$."

8. 4,293,372 "Growth of Single-Crystal Magnetoplumbile."

The last two patents were based on work performed under the present contract. The other patents were based on work performed under the previous contracts.